COEXISTING BIOTITES AND HORNBLENDES FROM SOME GRANITIC ROCKS IN SOUTHERN KITAKAMI MOUNTAINS, JAPAN

SATOSHI KANISAWA

*Department of Earth Science, College of Arts and Sciences, Tohoku
University, Sendai*

Coexisting biotites and hornblendes in granitic rocks from the Kitakami mountains were chemically analyzed. Most of biotites have composition near the phlogopite-annite series. Hornblendes of the Hitokabe, Tono and Kesengawa masses are similar in composition one another, but those of the Hirota mass are poorer in Al and Na+K and richer in Si than those of the former three masses. The Mg-Fe⁺² distribution coefficients for coexisting biotite-hornblende pairs from the Hitokabe, Tono, Kesengawa and Hirota masses are 0.79, 0.69, 0.70 and 0.67, respectively. These differences depend largely upon Al contents in these minerals.

1. INTRODUCTION

Recently, Onuki (1969) has summarized geologic features of the Kitakami mountains, and Katada *et al.* (1971) proposed zonal arrangement of the Cretaceous granitic rocks of the district on the basis of field occurrence, petrography and petrochemistry.

In the Kitakami mountains, large amounts of sedimentary and effusive rocks belonging to the Paleozoic and the Mesozoic in age are exposed, and are intruded by many kinds of plutonic rocks. Almost all granitic rocks of the district belong to the late Mesozoic and their K-Ar ages are 105-129 m.y., according to Kawano and Ueda (1965). These Cretaceous granitic rocks of the district have following peculiar characteristics among the Japanese granitic rocks. They have scarcely suffered crustal move-

ments after their intrusion, and they have not been covered so widely by the younger sediments, thus their original features and rock facies at the period of intrusion might have been almost maintained. Moreover, these granitic rocks form many bodies of various sizes, comprising various rock facies.

In the present paper, the chemical features of biotites and hornblendes from granitic rocks in the southern part of the Kitakami mountains are described.

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2. PETROGRAPHY OF ROCKS

Biotites and hornblendes were separated and chemically analyzed from four independent granitic masses; the Hitokabe,

the Tono, the Kesengawa and the Hirota masses. The Hitokabe mass consists of typical hornblende-biotite granodiorite and hornblende-biotite-quartz diorite and their data were already presented (Kanisawa, 1969). The Tono mass is composed of biotite granodiorite, biotite-hornblende granodiorite and hornblende-biotite-quartz diorite, sometimes containing clinopyroxene. The Kesengawa mass is composed of biotite-hornblende granodiorite and biotite-hornblende-quartz diorite, sometimes containing clinopyroxene, and contain abundant dark inclusions of various size. The Hirota mass

consists of clinopyroxene-biotite-hornblende granodiorite. From the facts that the effect of contact metamorphism is limited in narrow part, rock facies is fine to medium grained and massive, and optical properties of plagioclases scatter in wider range than those of the other masses (Kanisawa, 1972), the Hirota mass may represent a shallow intrusive body.

Distribution of granitic rocks and locations of the analyzed samples are shown in Fig. 1.

Chemical analyses of granitic rocks containing analyzed minerals are given in

Fig. 1. Distribution of granitic rocks of the Kitakami mountains, with locations of the analyzed samples, I; Ichinoseki, T; Tono, O; Ofunato, K; Kamaishi.

Table 1. From these data, granitic rocks of the district show the trend of typical calc-alkaline rock series. No distinct differences in chemical composition are recognized

among these granitic masses, though the Hirota mass is slightly richer in K_2O than in others. Thus, normative Q -Or-(Ab+An) diagram shows that all analyzed rocks fall

Table 1. Chemical analyses and norms of some analysed hornblende and biotite bearing granitic rocks.

	Tono				Kesengawa		Hirota			
	TN-1	TN-2	TN-3	TN-4	KS-1	KS-2	HR-1	HR-2	HR-3	HR-4
SiO ₂	56.52	62.34	67.01	70.35	57.11	64.97	60.60	61.56	62.77	62.53
TiO ₂	1.11	0.58	0.41	0.25	0.90	0.59	0.73	0.71	0.67	0.68
Al ₂ O ₃	17.71	16.74	15.17	15.54	17.54	15.33	15.62	15.74	15.09	16.17
Fe ₂ O ₃	1.84	2.46	2.22	1.99	3.17	2.32	2.62	2.44	2.32	2.07
FeO	5.48	3.14	2.29	0.90	4.41	2.78	4.18	3.89	3.62	3.22
MnO	0.13	0.09	0.07	0.06	0.11	0.09	0.11	0.10	0.10	0.12
MgO	3.92	2.73	1.79	0.76	3.25	1.99	3.03	2.87	2.67	2.13
CaO	7.09	5.65	3.91	3.23	6.52	4.31	5.72	5.75	5.08	4.87
Na ₂ O	3.25	3.19	3.37	4.85	3.64	3.40	3.10	2.78	2.83	3.58
K ₂ O	1.86	1.63	2.70	1.73	2.40	3.21	3.17	3.33	3.62	3.15
H ₂ O ₊	0.98	0.71	0.63	0.43	1.05	1.14	0.79	0.71	0.67	0.85
H ₂ O ₋	0.05	0.04	0.04	0.04	0.04	0.07	0.05	0.01	0.03	0.20
P ₂ O ₅	0.22	0.14	0.16	0.08	0.23	0.16	0.17	0.18	0.16	0.24
Total	100.16	99.44	99.77	100.21	100.37	100.36	99.89	100.07	99.63	99.81
Q	8.13	20.57	25.67	27.75	8.08	20.81	13.28	16.15	17.71	16.08
C	—	—	—	0.01	—	—	—	—	—	—
F { or	11.01	9.62	15.96	10.23	14.18	18.96	18.74	19.68	21.41	18.63
	ab	27.46	26.99	28.51	40.98	30.76	28.72	26.20	23.48	23.89
	an	28.13	26.49	18.24	15.48	24.41	17.07	19.29	20.60	17.74
Di { wo	2.37	0.27	0.07	—	2.70	1.38	3.33	2.82	2.70	1.64
	en	1.33	0.17	0.05	—	1.68	0.87	1.98	1.69	1.63
	fs	0.94	0.08	0.01	—	0.86	0.42	1.17	0.98	0.92
Hy { en	8.47	6.66	4.43	1.90	6.45	4.11	5.60	5.49	5.05	4.37
	fs	5.99	2.85	1.81	—	3.33	1.95	3.33	3.14	2.88
Mt	2.67	3.57	3.22	2.37	4.59	3.36	3.80	3.55	3.36	2.99
Il	2.11	1.11	0.78	0.47	1.72	1.12	1.38	1.35	1.28	1.29
Hm	—	—	—	0.35	—	—	—	—	—	—
Ap	0.50	0.34	0.37	0.20	0.54	0.37	0.40	0.44	0.37	0.57
D.I.	46.60	57.18	70.14	78.96	53.02	68.49	58.22	59.31	63.01	64.94
Mg/Mg+Fe ⁺² +Mn	0.58	0.66	0.64	0.59	0.61	0.63	0.65	0.63	0.63	0.62

TN-1. Clinopyroxene bearing hornblende-biotite-quartz diorite.

TN-2. Hornblende-biotite granodiorite.

TN-3. Hornblende-biotite granodiorite.

TN-4. Biotite granodiorite.

KS-1. Hornblende-biotite-quartz diorite.

KS-2. Clinopyroxene bearing hornblende-biotite granodiorite.

HR-1. ~ HR-4. Clinopyroxene-hornblende-biotite granodiorite.

Analyst: S. Kanisawa

Fig. 2. Normative Q-Or-(Ab+An) diagram of host rocks containing the analyzed minerals. The same symbols are used in all figures.

within the granodiorite field (Fig. 2).

3. OCCURRENCES OF BIOTITE AND HORNBLENDE

Biotites are generally anhedral and show pleochroism with X =pale yellow and Z =brown to light brown. Those of the Hirota mass are somewhat reddish brown. Sometimes biotites are altered to chlorite and sphene. Apatite, zircon and iron ore are common inclusions. Biotites from the Hirota mass are often rimmed with small needle crystals of iron ore.

Hornblendes are generally subhedral prismatic crystals and green to brownish green in Z -axial colour. Simple twinning on (100) is common. Sometimes, hornblendes from the Tono mass show weak zonal structure having brownish core and greenish rim. Inclusions of plagioclase and biotite are common, and rarely clinopyroxene occurs as core of hornblende. Rocks of the Hirota mass always contain clinopyroxene core having $b=Y$ and c -axis in common with the associated hornblende, which may have been

produced by reaction of granitic magma with pre-existing clinopyroxene during crystallization. Frequently, hornblendes of the Hirota mass have irregular actinolitic part showing $2Vx=73-75^\circ$, $c\wedge Z=16^\circ$, and very weak pleochroism. Apatite and iron ore are common inclusions in hornblendes of all the masses. Usually, biotite and hornblende are closely associated with sharp boundary, both as inclusions and contiguous grains, but they do not show any reaction relation with each other. Though the sequence of crystallization is uncertain, they may have crystallized almost simultaneously, or biotite may have been slightly later than hornblende. In all analyzed rocks, K-feldspar and magnetite coexist with biotite and hornblende.

4. CHEMICAL COMPOSITION OF MINERALS

In total ten specimens of biotite and nine of hornblende were separated from the host rocks and chemically analysed. These biotites and hornblendes are all coexisting pairs except one biotite (TN-4) of the Tono mass which is not associated with hornblende. All analyses were made by the conventional wet method, the alkalis being determined by means of atomic absorption spectroscopy, and TiO_2 and MnO by colorimetry. Chemical composition and structural formulae of analysed minerals are given in Tables 2 and 3. The data of the Hitokabe granodioritic mass of the district (Kanisawa, 1969) is included here for discussion. Structural formulae were calculated on the basis of 24 (O, OH) from the analytical data.

Biotites are all well within trioctahedral biotite field defined by Deer *et al.* (1963b). Their SiO_2 content is within 36.4–38.8% and other components also vary only in very narrow ranges. Their main variations in-

Table 2. Chemical analyses of biotites from some granitic rocks, Kitakami Mountains.

	Tono				Kesengawa		Hirota			
	TN-1	TN-2	TN-3	TN-4	KS-1	KS-2	HR-1	HR-2	HR-3	HR-4
SiO ₂	38.84	36.44	36.42	36.40	36.63	36.55	37.61	38.09	37.40	36.97
TiO ₂	3.16	3.70	3.34	3.94	3.79	3.86	4.33	4.35	4.70	5.02
Al ₂ O ₃	13.49	14.00	14.69	14.88	14.24	13.85	13.19	12.63	12.64	12.98
Fe ₂ O ₃	2.19	3.52	3.17	3.91	2.77	3.84	2.27	3.64	2.44	1.04
FeO	18.11	16.32	16.29	12.98	17.51	15.95	16.68	16.50	17.48	17.91
MnO	0.27	0.27	0.40	0.85	0.33	0.45	0.31	0.25	0.28	0.34
MgO	9.78	11.88	11.85	13.50	10.80	11.22	11.82	10.62	11.32	12.15
CaO	1.75	0.31	0.45	0.24	1.37	1.51	1.34	1.39	1.13	0.63
Na ₂ O	0.35	0.27	0.37	0.22	0.33	0.33	0.36	0.72	0.36	0.31
K ₂ O	7.78	8.57	8.74	8.46	8.18	7.60	8.25	8.18	8.44	8.66
H ₂ O ⁺	3.90	4.18	3.13	4.08	3.39	4.10	3.71	2.85	3.41	3.44
H ₂ O ⁻	0.02	0.02	0.03	0.11	0.10	0.11	0.14	0.10	0.17	0.04
Total	99.64	99.48	99.87	99.57	99.44	99.37	100.01	99.32	99.77	99.49
Numbers of ions on the basis of 24(O,OH)										
Si	5.867	5.509	5.607	5.439	5.616	5.530	5.682	5.864	5.717	5.653
Al ^{IV}	2.133	2.491	2.393	2.561	2.384	2.466	2.318	2.136	2.273	2.336
Al ^{VI}	0.265	—	0.268	0.055	0.184	—	0.026	0.151	—	—
Ti	0.358	0.420	0.386	0.442	0.435	0.438	0.490	0.503	0.539	0.576
Fe ⁺³	0.248	0.399	0.366	0.437	0.318	0.436	0.257	0.421	0.281	0.119
Fe ⁺²	2.280	2.056	2.090	1.615	2.237	2.011	2.100	2.117	2.227	2.282
Mn	0.034	0.034	0.052	0.108	0.042	0.057	0.040	0.032	0.036	0.044
Mg	2.216	2.694	2.737	3.026	2.484	2.546	2.679	2.453	2.596	2.787
Ca	0.283	0.050	0.074	0.039	0.225	0.245	0.217	0.229	0.185	0.103
Na	0.102	0.080	0.111	0.063	0.098	0.096	0.105	0.214	0.106	0.092
K	1.501	1.654	1.718	1.614	1.600	1.469	1.592	1.607	1.647	1.690
OH	3.929	4.212	3.213	4.065	3.464	4.136	3.737	2.925	3.474	3.506
Mg/Mg + Fe ⁺² + Mn	0.489	0.563	0.561	0.637	0.522	0.552	0.556	0.533	0.534	0.545

Analyst: S. Kanisawa

clude differences in contents of Mg⁺², Fe⁺², Mn⁺² and Ti⁺⁴, and differences in the amount of Al in tetrahedral co-ordination. Mg/Fe ratios of biotites in the Tono mass tend to increase with increasing differentiation index (D.I.) of the host rocks, and similar tendency is shown in the Kesengawa mass. Al₂O₃ content ranges from 13 to 15% in the Tono, Kesengawa and Hitokabe masses, but that in the Hirota mass is somewhat lower, being about 12%, whereas TiO₂ content of biotites in the Hirota mass is higher than that of the other masses. From the com-

parison of the structural formulae, biotites of the Hirota mass have no or little octahedral Al. According to Nockolds (1947), biotites associated with pyroxenes in calc-alkaline igneous rocks are poor in Al and rich in Ti, thus octahedral sites in biotite are frequently occupied by Ti. Biotites in the Hirota mass are always associated with clinopyroxene and poor in Al and rich in Ti, thus their chemical characteristics agree with Nockolds' opinion. Similar biotites rich in Ti and poor in Al occur in the Orikabe granodiorite mass in the Kitakami moun-

Table 3. Chemical analyses of hornblendes from some granitic rocks, Kitakami Mountains.

	Tono			Kesengawa		Hirota			
	TN-1	TN-2	TN-3	KS-1	KS-2	HR-1	HR-2	HR-3	HR-4
SiO ₂	46.76	46.54	46.23	45.61	46.10	47.97	47.86	48.10	48.79
TiO ₂	1.23	1.59	1.45	1.66	1.57	1.13	1.25	1.13	1.22
Al ₂ O ₃	7.26	6.96	8.66	8.18	7.48	4.78	5.33	5.75	4.14
Fe ₂ O ₃	3.82	4.73	3.61	4.00	4.88	4.35	3.94	3.31	3.49
FeO	14.00	11.63	11.96	12.84	11.95	11.96	12.55	13.07	13.57
MnO	0.38	0.45	0.52	0.43	0.60	0.51	0.50	0.49	0.68
MgO	10.90	11.63	12.14	11.49	11.76	12.78	12.56	12.71	12.83
CaO	12.35	12.82	11.63	11.83	11.75	13.22	12.23	11.71	11.35
Na ₂ O	1.10	1.35	1.36	1.21	1.38	1.10	1.19	1.16	1.12
K ₂ O	0.82	0.68	0.76	0.85	0.87	0.60	0.72	0.67	0.58
H ₂ O ₊	1.91	2.06	1.81	1.99	1.99	1.87	1.92	2.17	2.17
H ₂ O ₋	0.02	0.00	0.04	0.00	0.07	0.06	0.04	0.09	0.05
Total	100.40	100.55	100.17	100.09	100.40	100.33	100.09	100.36	99.99
Numbers of ions on the basis of 24(O, OH)									
Si	6.927	6.830	6.811	6.757	6.810	7.090	7.079	7.062	7.200
Al ^{IV}	1.073	1.117	1.189	1.243	1.190	0.832	0.921	0.938	0.719
Al ^{VI}	0.193	0.084	0.312	0.183	0.109	—	0.007	0.056	—
Ti	0.137	0.175	0.160	0.185	0.174	0.125	0.138	0.124	0.135
Fe ⁺³	0.425	0.521	0.400	0.444	0.541	0.482	0.437	0.365	0.386
Fe ⁺²	1.728	1.422	1.468	1.585	1.471	1.473	1.547	1.599	1.669
Mn	0.048	0.055	0.065	0.054	0.075	0.064	0.062	0.061	0.085
Mg	2.422	2.822	2.683	2.557	2.606	2.833	2.786	2.799	2.840
Ca	1.960	1.845	1.836	1.877	1.860	2.094	1.938	1.842	1.795
Na	0.315	0.384	0.387	0.347	0.395	0.314	0.341	0.329	0.321
K	0.155	0.127	0.143	0.160	0.165	0.114	0.137	0.125	0.110
OH	1.886	2.014	1.779	1.966	1.961	1.843	1.894	2.125	2.136
Mg/Mg + Fe ⁺² + Mn	0.577	0.656	0.636	0.609	0.628	0.648	0.634	0.628	0.618

Analyst: S. Kanisawa

tain (Kato, 1972). Biotites having no octahedral aluminum in the Tono and Kesengawa masses contain OH exceeding 4.1. The amounts of (Fe⁺² + Mg) are almost constant (4.5–4.8), thus Fe⁺² and Mg are mutually compensative. The value of Ti + Fe⁺³ + Al^{IV} is approximately 1, and Si ranges from 5.43 to 5.86, thus the compositions of biotites are plotted near the phlogopite-annite series in the diagram of Deer *et al.* (1963b) (Fig. 3). With increasing differentiation index of the host rocks, the amount of Si in the biotite decreases (Fig. 4), thus

the composition of biotites change toward siderophyllite-eastonite series as shown in Fig. 3.

The compositions of hornblendes is also not so variable, but those of the Hirota mass are richer in SiO₂ and poorer in Al₂O₃ and TiO₂ than those of the other masses. According to Leake's classification (Leake, 1968), all hornblendes from the Hitokabe mass fall within ferro-hornblende field, while those from other masses within magnesio-hornblende field. In hornblendes from the Tono and Kesengawa masses, the Mg/Fe

Fig. 3. Variation diagram of biotites after Deer *et al.* (1963b).

Fig. 4. Relationship between Si content in biotites and the differentiation index of the host rocks.

ratio tend to increase with increasing D.I. of the host rocks as in the case with the biotites. From structural formulae and the atomic ratios plotted on the diagram of Deer *et al.*, (1963a), it is evident that hornblendes of the Hitokabe, Tono and Kesengawa masses are similar in composition to one another, while those of the Hirota mass are poorer in Al^{IV} , $Na+K$, and $Al^{VI}+Ti+Fe^{+3}$ and richer in Si than those of the other masses (Fig. 5). Tetrahedral Al substituting Si is more than one atom per unit formula in

Fig. 5. Al^{IV} - $Na+K$ diagram and Al^{IV} - $Al^{VI}+Fe^{+3}+Ti$ diagram of hornblendes after Deer *et al.* (1963a).

hornblendes from the Tono, Hitokabe and Kesengawa masses, but less than one in those from the Hirota mass. Similar hornblendes poor in Al occur as the reaction products of primary pyroxene in shallow intrusives accompanying cauldron subsidence such as the Ben Nevis complex (Haslam, 1968) and the Tamagawa plutonic complex (Murakami, 1969). Generally, in hornblendes from each mass in this district, increasing amounts of tetrahedral Al are balanced both by increased entry of alkalis into the A site and by substitution of tri-

Fig. 6. Relationship between $\text{Mg}/(\text{Mg}+\text{Fe}^{+2}+\text{Mn})$ of the minerals and those of their host rocks.

and quadri-valent cations in the octahedral Y site. Ca per formula unit is 1.8–2.0 in all granitic masses in this district.

The $\text{Mg}/(\text{Mg}+\text{Fe}^{+2}+\text{Mn})$ ratios of biotites and hornblendes are practically correlated with those of their host rocks as shown in Fig. 6.

The relation of Fe^{+3} – Fe^{+2} –Mg contents in coexisting biotites and hornblendes is shown in Fig. 7. It is clear that all hornblendes are richer in Fe^{+3} than coexisting biotites, and that the composition of most of biotites is defined by oxygen fugacities equal to or slightly higher than those of Ni–NiO buffer estimated by Wones and Eugster (1965). All biotites associate with K-feldspar and magnetite, thus the oxygen fugacities of granitic magmas in the Kitakami mountains were approximately constant. However, in the Tono granitic mass, oxygen fugacity increased slightly with cooling, therefore, biotites tend to be slightly rich in Mg with increasing D.I. of the

host rocks.

Fig. 7. Fe^{+3} – Fe^{+2} –Mg diagram of coexisting biotites and hornblendes. Large symbols are biotites and small symbols are hornblendes. Dashed lines represent compositions of 'buffered' biotites in the ternary system $\text{KFe}_3^{+3}\text{AlSi}_3\text{O}_{12}\text{H}_{-1}$ – $\text{KFe}_3^{+2}\text{AlSi}_3\text{O}_{10}(\text{OH})_2$ – $\text{KMg}_3\text{AlSi}_3\text{O}_{10}(\text{OH})_2$ estimated by Wones & Eugster (1965).

Table 4. Mg/Fe^{+2} , $Mg/Mg+Fe^{+2}$ and Mn/Mg ratios and K_D (Mg/Fe^{+2})_(bio/hor) and K_D (Mn/Mg)_(bio/hor) for biotites and hornblendes in granitic rocks from the Kitakami mountains.

	No.	Biotite			Hornblende			$K_D(Mg/Fe^{+2})$	Aver.	$K_D(Mn/Mg)$	Aver.
		Mg/Fe^{+2}	$Mg/Mg+Fe^{+2}$	Mn/Mg	Mg/Fe^{+2}	$Mg/Mg+Fe^{+2}$	Mn/Mg				
Hitokabe	1*	0.980	0.495	0.020	1.203	0.547	0.036	0.81		0.56	
	2	1.000	0.500	0.020	1.321	0.569	0.032	0.76		0.63	
	3	1.130	0.531	0.021	1.383	0.580	0.038	0.82	0.79	0.55	0.62
	6	0.923	0.480	0.020	1.218	0.549	0.030	0.76		0.67	
	9	0.958	0.489	0.028	1.189	0.543	0.040	0.81		0.70	
Tono	TN-1	0.972	0.483	0.016	1.402	0.584	0.020	0.69		0.80	
	TN-2	1.310	0.567	0.013	1.985	0.664	0.020	0.66	0.69	0.65	0.72
	TN-3	1.309	0.567	0.019	1.827	0.646	0.024	0.72		0.79	
Kesengawa	KS-1	1.110	0.526	0.017	1.614	0.617	0.021	0.69		0.81	
	KS-2	1.266	0.559	0.022	1.771	0.639	0.029	0.71	0.70	0.76	0.79
Hirota	HR-1	1.275	0.561	0.015	1.924	0.658	0.023	0.66		0.65	
	HR-2	1.158	0.537	0.013	1.801	0.643	0.022	0.64		0.59	
	HR-3	1.166	0.538	0.014	1.751	0.636	0.022	0.67	0.67	0.64	0.60
	HR-4	1.222	0.550	0.016	1.702	0.630	0.030	0.72		0.53	

* Numbers of the Hitokabe mass are the same as Kanisawa's (1969) Tables 2 and 3.

5. DISTRIBUTION OF Fe^{+2} , Mg AND Mn IN COEXISTING BIOTITES AND HORNBLENDES

Most of the granitic rocks in the present district have coexisting biotites and hornblendes. It is ambiguous whether these two minerals were equilibrated or not in the host rocks, but the fact that they do not show any reaction texture with each other suggests the possibility that they have crystallized simultaneously. Biotites and hornblendes are assumed at present to have attained equilibrium to some extent.

The distribution coefficients for Mg , Fe^{+2} and Mn in coexisting biotite and hornblende are expressed as

$$K_D(\text{Mg}/\text{Fe}^{+2})_{(\text{bio}/\text{hor})} = (\text{Mg}/\text{Fe}^{+2})_{\text{bio}} \div (\text{Mg}/\text{Fe}^{+2})_{\text{hor}}, \text{ and } K_D(\text{Mn}/\text{Mg})_{(\text{bio}/\text{hor})} = (\text{Mn}/\text{Mg})_{\text{bio}} \div (\text{Mn}/\text{Mg})_{\text{hor}}.$$

The Mg/Fe^{+2} , $\text{Mg}/\text{Mg} + \text{Fe}^{+2}$ and Mn/Mg ratios and K_D of coexisting biotite-hornblende pairs are shown in Table 4. It is clear from these data that K_D values for hornblende-biotite pairs are approximately constant in each granitic mass, that is, $K_D = 0.79$ in the Hitokabe mass, 0.70 in the Kesengawa mass, 0.69 in the Tono mass and 0.67 in the Hirota mass. The distribution of Mg/Fe^{+2} between biotites and hornblendes

in each granitic mass is shown in Fig. 8.

The average K_D values for coexisting biotite-hornblende in granitic rocks from various districts are given below; $K_D = 0.83$ and 0.82 in the Garabal Hill-Glen Fyne complex and the Morvern-Strontian complex respectively (Nockolds and Mitchell, 1948), $K_D = 0.82$ in the Southern California batholith (Larsen and Draisin, 1950), $K_D = 0.77$, 0.81 and 0.64 in the rocks of the youngest age, the intermediate age and the oldest age from the Central Sierra Nevada batholith, respectively (Dodge *et al.*, 1968 and 1969), $K_D = 0.65$ in the zoned plutons of Northern Sierra Nevada (Hietanen, 1971), $K_D = 0.56$ in the Ben Nevis complex (Haslam, 1968), $K_D = 0.92$ in the granitic rocks near the San Andreas Fault (Dodge and Ross, 1971), $K_D = 0.79$ in the rocks of amphibolite-quartz diorite series from the Takanuki district (Ogura, 1959), $K_D = 0.64$ in the Orikabe granodioritic mass, Kitakami mountains (Kato, 1972), and $K_D = 0.93$ in the Ryoke granitic rocks of the Takato district, Central Japan (Kanisawa, unpublished data). Mueller (1963) posed an interesting question whether equilibrium can be established or not with respect to Fe^{+2} and Mg when the coexisting plagioclase is zoned. According

Fig. 8. Distribution of $\text{Mg}-\text{Fe}^{+2}$ between coexisting biotite and hornblende.

to Mueller, existence of such a plagioclase showing compositional zoning indicates kinetic situation unfavourable for equilibrium. Biotite-hornblende pairs of granitic rocks from the Kitakami mountains are all associated with strongly zoned plagioclase, however, K_D values for biotite-hornblende pairs are approximately constant in each granitic mass in the Kitakami mountains and other districts. This fact shows that biotite-hornblende pairs were equilibrated on the Mg-Fe⁺² distribution to some extent even in condition that zoned plagioclase crystallized in granitic magma.

Distribution coefficients of Mg-Fe⁺² for biotite-hornblende pairs from areas of different metamorphic facies do not show any systematic variation with the difference of metamorphic facies (Leelanandam, 1970). The K_D values for biotite-hornblende pairs from granitic rocks are also difficult to correlate simply with PT-condition. The variation in K_D values for coexisting biotite-hornblende pairs either from magmatic or from metamorphic rocks, could be influenced largely by the differing mineralogy and

chemical composition of the host rocks, the multivariant nature of the minerals, and to the chemicommechanical effects consequent on the regional and environmental influences characteristic of individual terrains (Mueller; 1963, Saxena; 1968, Leelanandam; 1970). K_D values of Mg-Fe⁺² for coexisting biotite-hornblende pairs in granitic rocks from the Kitakami mountains depend largely upon Al contents in these minerals, because biotites and hornblendes from the Hitokabe mass rich in tetrahedral Al have larger K_D value, but those from the Hirota mass poor in tetrahedral and octahedral Al have smaller K_D value.

Mn-Mg distribution between coexisting biotite-hornblende pairs in granitic rocks from the Kitakami mountains are indicated in Table 4 and Fig. 9. $K_D(\text{Mn/Mg})$ values vary from 0.81 to 0.53 and average value in the Kesengawa mass is the largest, while that in the Hirota mass is the smallest. Recently, Greenland *et al.* (1968) has shown that the distribution of Mn between coexisting biotite and hornblende reflects exchange at subsolidus temperatures rather than initial crystallization temperatures and that

Fig. 9. Distribution of Mn-Mg between coexisting biotite and hornblende.

there is a positive correlation of K_D with TiO_2 content of biotite for biotite-hornblende pairs from the Southern California and Sierra Nevada batholiths. However, in the granitic rocks of the district, it is difficult to investigate whether the concepts of Greenland *et al.* can be applicable or not, and the K_D (Mn/Mg) values for biotite-hornblende pairs is independent of TiO_2 content of biotite.

6. SUMMARY

Biotites and hornblendes in the Hitokabe, Tono, Kesengawa and Hirota masses were chemically analyzed. Compositions of most biotites are rich in Si, thus they are close to phlogopite-annite series. Generally, biotites become richer in siderophyllite-eastonite molecule with increasing differentiation index of their host rocks. Biotites in the Hirota mass are rich in TiO_2 and poor in Al_2O_3 . Hornblendes of the Hitokabe, Tono and Kesengawa masses are similar in composition one another, while those of the Hirota mass are poorer in Al^{IV} , $Na+K$, and $Al^{VI}+Ti+Fe^{+3}$ and richer in Si than those of the other masses. Generally, substitution of Si by tetrahedral Al in hornblendes of granitic rocks from the district are balanced by $Na+K$ in the A site and Fe^{+3} , Al^{VI} and Ti ions in the Y site. The Mg-Fe distribution coefficients for coexisting biotite-hornblende pairs are approximately constant in each granitic mass, that is, K_D (Mg/ Fe^{+2}) for biotite/hornblende is 0.79 in the Hitokabe mass, 0.70 in the Kesengawa mass, 0.69 in the Tono mass, and 0.67 in the Hirota mass. The variation of K_D among each granitic mass depends largely upon Al contents in these minerals.

REFERENCES

- Deer, W.A., Howie, R.A. and Zussman, J. (1963a), Rock forming minerals, vol. 2; Chain silicates, Longmans, London.
- , ———, and ——— (1963b), Rock forming minerals, vol. 3; Sheet silicates, Longmans, London.
- Dodge, F.C.W., Papike, J.J. and Mays, R.E. (1968), Hornblendes from granitic rocks of the Central Sierra Nevada batholith, California, *Jour. Petrol.*, **9**, 378–410.
- , Smith, V.C. and Mays, R.E. (1969), Biotites from granitic rocks of the Central Sierra Nevada batholith, California, *Jour. Petrol.*, **10**, 250–271.
- , and Ross, D.C. (1971), Coexisting hornblendes and biotites from granitic rocks near the San Andreas fault, California, *Jour. Geol.*, **79**, 158–172.
- Greenland, L.P., Gottfried, D. and Tilling, R.I. (1968), Distribution of manganese between coexisting biotite and hornblende in plutonic rocks, *Geochim. Cosmochim. Acta*, **32**, 1149–1163.
- Haslam, H.W. (1968), The crystallization of intermediate and acid magmas at Ben Nevis, Scotland, *Jour. Petrol.*, **9**, 84–104.
- Hietanen, A. (1971), Distribution of elements in biotite-hornblende pairs and in an orthopyroxene-clinopyroxene pair from zoned plutons, Northern Sierra Nevada, California, *Contrib. Mineral. Petrology*, **30**, 161–176.
- Kanisawa, S. (1969), On the Hitokabe granodioritic mass, Kitakami Mountainland, *Jour. Japan. Assoc. Min. Petr. Econ. Geol.*, **62**, 275–288 (in Japanese with English abstract).
- , (1972), Optical properties of plagioclases in some granitic rocks, Kitakami mountains, Northern Japan, *Jour. Geol. Soc. Japan*, **78**, 301–307.
- Katada, M., Onuki, H., Kato, Y., Kanisawa, S., Ono, C. and Yoshii, M. (1971), Zonal arrangement of the Cretaceous granitic rocks, Kitakami mountainland, *Jour. Japan. Assoc. Min. Petr. Econ. Geol.*, **65**, 230–245 (in Japanese with English abstract).
- Kato, Y. (1972), Petrology of the Orikabe granitic body, Kitakami mountainland, *Jour. Japan. Assoc. Min. Petr. Econ. Geol.*, **67**, 50–59, (in Japanese with English abstract).
- Kawano, Y. and Ueda, Y. (1965), K-A dating on the igneous rocks in Japan (II) — Granitic rocks in Kitakami massif, *Jour. Japan. Assoc. Min. Petr. Econ. Geol.*, **53**, 143–154 (in Japanese with English abstract).
- Larsen, E.S., Jr. and Draisin, W.M. (1950), Composition of the minerals in the rocks of the Southern California batholith, *Rep. 18th Int. Geol. Congr. Great Britain*, 1948, pt. 2, 66–79.

- Leake, B.E. (1968), A catalog of analyzed calciferous and subcalciferous amphiboles together with their nomenclature and associated minerals, *Geol. Soc. Am. Special Paper*, no. 98.
- Leelanandam, C. (1970), Chemical mineralogy of hornblendes and biotites from the charnockitic rocks of Kondapalli, India, *Jour. Petrol.*, **11**, 475-505.
- Mueller, R.F. (1963), Interaction of chemistry and mechanics in magmatism, *Jour. Geol.*, **71**, 759-772.
- Murakami, N. (1969), Two contrastive trends of evolution of biotite in granitic rocks, *Jour. Japan. Assoc. Min. Petr. Econ. Geol.*, **62**, 223-248.
- Nockolds, S.R. (1948), The relation between chemical composition and paragenesis in the biotite micas of igneous rocks, *Am. Jour. Sci.*, **245**, 401-420.
- , and Mitchell, R.L. (1948), The geochemistry of some Caledonian plutonic rocks: a study in the relationship between the major and trace elements of igneous rocks and their minerals, *Trans. Royal Soc. Edinb.*, **61**, 533-575.
- Ogura, Y. (1959), On the granitization of some basic rocks of the Gosaisho-Takanuki district, Southern Abukuma plateau, Japan, *Jour. Geol. Geogr.*, **29**, 171-198.
- Onuki, Y. (1969), Geology of the Kitakami Massif, Northeast Japan, *Tohoku Univ. Inst. Geol. Pal. Contr.*, no. 69, 1-239 (in Japanese with English abstract).
- Saxena, S.K. (1968), Distribution of elements between coexisting minerals and the nature of solid solution in garnet, *Am. Mineral.*, **53**, 994-1014.
- Wones, D.R. and Eugster, H.P. (1965), Stability of biotite: experiment, theory, and application, *Am. Mineral.*, **50**, 1228-1272.

南部北上山地の 2, 3 の花崗岩体中の共存する黒雲母と角閃石

蟹 沢 聰 史

北上山地の人首・遠野・気仙川および広田岩体中の共存する黒雲母と角閃石について化学分析を行なった。黒雲母は phlogopite-annite series に近い組成をもつものが大部分であるが、岩石の D.I. が大きくなるにつれて、siderophyllite-eastonite に近い組成に変わる。角閃石については、人首・遠野・気仙川の各岩体のものは互に似た化学組成を示すが、広田岩体のものは Al, Na+K にとばしく、Si にとむ。人首・遠野・気仙川・広田岩体中で共存する黒雲母と角閃石の $K_D(Mg/Fe^{+2})$ は各岩体ごとにほぼ一定で、それぞれ 0.79, 0.69, 0.70, および 0.67 である。K_D の小さい岩体の角閃石は一般に Al にとばしい。